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RETROSPECTIVE RECONSTRUCTION OF RADIONUCLIDE CONTAMINATION OF THE TECHA RIVER CAUSED BY LIQUID WASTE DISCHARGE FROM RADIOCHEMICAL PRODUCTION AT THE MAYAK PRODUCTION ASSOCIATION: 1949 - 1956

Joint Coordinating Committee on Radiation Effects Research
Project 1.3

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Background

Project 1.3 is a recently-approved pilot study designed to strengthen the source term information for the Techa River that is supporting JCCRER Project 1.1. The purpose of the feasibility study is to determine if the source term of radioactive materials released to the Techa River from the Mayak facility can be reconstructed from historical environmental measurements. This information is necessary for the correct estimation of the dose exposure of the population inhabiting near the banks of the Techa River in the 1950s and 1960s. The pilot study is investigating the feasibility of reconstructing the releases of a limited number of radionuclides from measurements made at a limited number of downriver locations, using historically measured water flow rates and total-beta radioactivity measurements, and considering the processes of radioactive decay and of sorption/desorption. A simple radionuclide mass balance approach is being used, based on a current successful technique for accounting for present-day observed migration of 90Sr. The feasibility study is evaluating its applicability to the complete Mayak operating period for radionuclides other than 90Sr.

The long-term objective of this project is reconstruction of the time-dependent source of radionuclides released to the waters, sediments, and environs of the Techa River from the facilities of the Mayak Production Association for the period beginning in 1949. The pilot study will indicate whether it is possible to determine the historical releases of a wider suite of radionuclides using the historical monitoring data from numerous locations along the river, rather than relying on a more uncertain reconstruction of quantities released at the point of discharge. Radionuclides to be considered include 90Sr, 106Ru, 137Cs, and 144Ce. Locations will include points on the upper, middle, and lower Techa River.

To determine the rate of input of radionuclides to the Techa River system, the following approach is being used. The Techa River is being depicted as a series of characteristic sites with controlled hydro-sections. For each site on the river, a system of recurrent (with time) equations has been compiled for radioactivity balance accounting for the radioactivity inflow at the inflowing section, activity discharge with water at outflowing section, and the reduction of activity because of radioactive decay. The equations change with time to account for the changing nature of the river regime. Effective (integral for each of the investigated segments of the river) sorption constants which characterize the rate of washing of the radionuclides in the river system components (flood-plain soil, bottom sediments) for 90Sr and 137Cs are being defined, based on the inventory of the stated radionuclides deposited at each of the studied river sites, and data on water concentration and radioactive removal. All the information on radioactive contamination of the river system components during the period 1949 - 1996 will be used. Solution of the series of equations generated will provide information on the rate of input of these radionuclides into the upper end of the river.

The effort for the feasibility study includes two stages to be performed over a period of 12 months.

Stage 1 (first three months)

The following activities have been performed.

Data has been collected, analyzed, and a computer data base prepared of the following information:

- monthly data on the water flow rates of the Techa River at different sites of the river (hydro-sections);
- data on the average monthly total beta-activity of the water in the Techa River components at the stated hydro-sections;
- experimental measurement data (1949 - 1965) on radionuclide contamination of water, bottom sediments and flood-plain soil at different sites of the Techa River;
- data on the change with time of the exposure dose rate and density of the beta-activity flux at different sites of the flood-plain;
- meteorological data (precipitation. temperature, wind speed etc.) with monthly normalization.

A brief description of the hydrographic and hydrological characterization of the Techa River has been prepared, including the main types of the soil and sediments composing the flood-plain and river bed, entitled "Hydrographic and Hydrological Description of the Techa River with the Analysis of the Properties of the Major Types of Flood-Plain Soils and Bottom Sediments."

Based on the results of this stage, the summary document "Analysis of the experimental information on water flow and radioactive contamination of the Techa River system components" has been prepared.

Stage two (fourth through twelfth months)

The modeling approach is based on the model described in Mokrov "Semi-empirical model of 90Sr transfer with the water of the Techa River", *Radiation Safety Problems Journal*, "Mayak" PA, No. 3, 1996. The model is a simple mass balance applied to a series of river segments in a recursive fashion with time. This model was developed for predicting the future transport of 90Sr from the Techa River. The model has five empirical parameters, which to date have been evaluated using 20 years worth of data for the radionuclide Sr90.

The following activities are being performed in this stage of the feasibility study:

combining and computer implementation of the semi-empirical balance model of radionuclide transfer with Techa River water for additional radionuclides;

a series of calculations with the aim of determining the numerical values of the semi-empirical model parameters for the radionuclides and locations used;

estimation of the quantities of radionuclides entering the Techa River at the point of release, with emphasis on the period of earliest operations;

comparative analysis of the calculated results with the experimental data obtained in 1949 through 1996;

recommendation on elaboration or improvement of the methods of retrospective reconstruction of sources of radionuclide contamination of the water of the Techa River, including an description of an approach to determining the overall uncertainty of the calculated results;

Generalization of all the obtained data and preparation of the final report.

A discussion of the status of these areas will be made at the April SRG meeting in Chelyabinsk.

Relationship To Other JCCRER Projects

Project 1.3 is directly related to JCCRER Projects 1.1 and 1.2. The objective of Project 1.1 is the preparation of individual radiation dose estimates for the Techa River epidemiology cohort defined in Project 1.2. In order to calculate the radiation doses in Project 1.1, information on the quantities of radioactive material released to the Techa River is needed. Project 1.3 exists in direct response to the need identified for Project 1.1.

Both Projects 1.1 and 1.3 use "Techa River Models." Although the basic concepts of hydrological modeling are common to both projects, the structure and aims of the modeling efforts are different and complementary. The purpose of modeling the Techa River in Project 1.1 is reconstruction of radionuclide concentrations in river water and sediment, and external dose rates, at arbitrary locations (where the people were). The inputs to this modeling include either a source term (preferred) or upstream environmental measurements, and information on river water flow rates and river geometry. The purpose of the modeling in Project 1.3 is reconstruction of the source term at the point of release. The inputs to this modeling include measurements of radionuclide concentration at a limited number of fixed points (where the time-series measurements are available), as well as information on river flow rates, meteorology, etc.

Information developed in Project 1.3 will be shared with Project 1.1. To date, communications between the projects has been acceptable. Minor discrepancies and misunderstandings of data interpretation have arisen, but discussions via e-mail have resolved all outstanding questions.

Summary of Progress and Achievements of Phase 1

The initial focus of the first phase of Project 1.3 was understanding of the physical situation of the Techa River. The purpose of the present work is the re-creation and analysis of the hydrological parameters and properties of the basic types of soils combining the bed and pre-bed part of the river in the time period from 1949 to 1963. This time period is characterized with the intense release of radionuclides into the water of the Techa River (1949-1956) and hydrotechnical projects in the upper stream of the Techa River (1956-1963).

A brief report, "Hydrographic and Hydrological Description of the Techa River with the Analysis of the Properties of the Major Types of Flood-Plain Soils and Bottom Sediments," has been prepared. This document provides a summary of available data on the geography, geology, and hydrography of the Techa River.

Hydrographic and hydrological descriptions and properties of the rivers Techa and Iset are given for the present moment and for 1949, when the liquid radioactive waste dumping at "Mayak" PA to the Techa River was started.

Data characterizing physical and chemical properties of the flood-plain soils and bottom sediments of the rivers are given. This information dates back to the 1950s.

Information on suspensions, run-off, and chemical composition of water in the rivers is presented for 1941-1963.

Information necessary for understanding of the radionuclide migration processes in the river system is described, such as the time-dependence of water run-off, and the influence of the upstream swamps.

This report will be available at the April SRG meeting in Chelyabinsk.

The second, and primary, focus of the first phase of Project 1.3 was collection and interpretation of available historical environmental radioactivity monitoring data. A large amount of information has been found. It is summarized in a report, "The analysis of the experimental information about water drain and radioactive contamination of Techa river system components." This report will also be available at the April SRG meeting in Chelyabinsk. This report provides a description of the historical processes that lead to the releases, describes the types of data found and the methods historically used for environmental sample collection and analysis, and collects the data into a coherent narrative description of Techa River contamination that can be used as a basis for modeling.

The history of the Techa River contamination has two main periods, each of which has several subperiods. During the initial major period from 1949 through late 1951, the Techa River was intentionally used as the receiver for contaminated waste water. After this time during the second major period, the bulk of radioactive discharges were routed to Lake Karachai, but there were still several instances of relatively large release into the river itself.

Beginning in early 1949, low-level liquid radioactive wastes and cooling waters were discharged directly into the Techa River. In 1950, in order to reduce the volume of material going to the high-level waste storage, a process for "decontamination" of high-level wastes was introduced, with a portion of the radioactivity directed to the tanks and a portion released to the river. In July 1951, it was discovered that this process did not work as intended, and that during this period high concentrations of radionuclides had been released into the river. Also during this time, cooling water from the high-level waste tanks was discharged into the Techa River at the same location as the process wastes. Leaks in the tank-cooling lines caused some of these discharges to be highly contaminated. These "wild releases" were unmonitored and unnoticed until September 1951. Over this period, about half of the total release to the Techa River resulted from the technological releases and about half from the wild releases. In late 1951, several activities were started to control the releases and to remediate the environmental contamination. The main process wastes were diverted into Lake Karachai on October 28, 1951. However, drainage water, cooling water, and low-level sanitary waters were still released to the river. Some additional tank cooling "wild releases" occurred through mid 1952.

The low- and intermediate-level liquid wastes that were intentionally discharged came from several sources.

Cooling waters became contaminated by radioactive substances as a result of corrosion in cooling jackets. The radioactivity of cooling water usually was small - about 10-7-10-8 Ci per liter.

Drainage waters were pumped out from deep wells to decrease underground water levels. The chemical structure was close to normal groundwater. Drainage waters became contaminated by radioactive substances in groundwater from the communication with observation wells for high-level waste tanks. Characteristic beta-activity of these waters is in the range from 10-8 up to 10-3 Ci per liter.

"**Ladder waters"** (trapnie water – so named because of the ladders used to enter shielded rooms) were generated during floor washing of industrial rooms. They were very heavily polluted by radioactive substances if liquidation of emergency spills of technological solutions was carried out. In ladder waters, the basic amount of activity (up to 90 %) was transferred mainly on suspensions, passing subsequently through reservoirs as bottom sediments. The typical beta-activity of ladder waters ranges from 10-6 up to 10-2 Ci per liter.

Shower and laundry waters were created by washing overalls and footwear, and during staff washing in shower-rooms. Overalls were washed with solutions of soda, nitric acid, and hexametaphosphate. For washing rubber footwear, paraffin contact was applied. These waters also contained soap. Radioactivity of shower and laundry waters was in the range of 10-7 to 10-4 Ci per liter.

Cooling water from vacuum pumps became contaminated by radioactive and chemical substances with aerosols from vacuum lines. Beta-activity levels were 10-5 - 10-4 Ci per liter.

Desorbtion waters were generated during washing of radioactive products from devices and canyons. These waters were usually strongly polluted with chemical substances, since for washing, solutions of potassium permanganate, caustic soda, nitric acid, oxalic acid and even hydrofluoric acid were applied. The beta-activity of waters is reduced on a measure of the device washing from 10-2 down to 10-7 Ci per liter.

Laboratory waters were formed by washing of utensils contaminated with radioactive substances, and by reagent and researched solutions discharge in the water drain. Beta-activity of laboratory waters was 10-2 - 10-6 Ci per liter.

The **condensates** of multi-cyclones were polluted with all chemical substances which were used. Specific beta-activity of condensates were in the range from 10-1 down to 10-3 Ci per liter.

The radiochemical structure of substances contained in discharged waters was determined by the radiochemical structure of those products with which they were associated. If the contamination was made by certain products having constant radiochemical structure, the radiochemical structure of the discharged water also had a constant structure. Ladder, laboratory and cooling waters, condensates, and decantates all were of relatively constant composition. In drainage waters, the relative enrichment by ruthenium was observed because of selective soil sorbtion. In shower/laundry waters, relative de-enrichment of strontium was observed because of loss in sediment.

Additional releases to the Techa River occurred during the unmonitored releases from the high-level waste tanks. These apparently began in early 1950, but were not discovered until mid 1951. Although the discharges of most waste streams were diverted to Lake Karachai, several additional instances of release via this mechanism were noted, the radiation monitoring service reported non-authorized discharges on October 9, 26 and 31, November 2, and December 12, 1951, and January 19 and May 2, 1952. These monitored discharges are believed to have been smaller than the unmonitored earlier ones.

For the entire period of Techa river monitoring, the methods of river sampling have remained essentially unchanged. The water samples have always been sampled in clean glass or polyethylene utensils. At the sampling point, the utensils (bottle, retort, measured glass) have been washed in river water, and then immersed to a depth of 10-15 cm for water sampling. Volume of the water samples depends on the kind of analyses and ranges from 5 to 80 liters. The sampling frequency changed over various periods of time. In the period from 1952 to 1956, sampling at the point of water discharge (below hydraulic engineering structures) was carried out daily, and at other locations on the river (Techa-Brod, Staroe Asanovo, Nadyrov Most, Muslumovo, Brodokalmak, Russkaya Techa, Bugaevo, Pershino and Zatechenskoe), one or two times per month. In the period from 1960 to 1965, regular water sampling was carried out in Muslumovo and Pershino with a frequency of from once per day (during spring high waters or intensive high waters) to 5-15 times per month (during summer low waters periods). Since middle of the 1960s, the sampling in Muslumovo and Pershino was performed with cumulative samples. One-third of a liter per day was sampled and merged into a polyethylene canister. The resulting monthly average water sample was transferred to the laboratory for radiometric measurements. The bottom sediments were sampled by means of special sampling tools, which design have not undergone major changes for the entire period of field work. The sampling tool is a metal (in the 1950s - glass) tube, the bottom end of the tube has a pointed cutting edge.

The methods used to determine the levels and types of contamination were being developed in the early years of Mayak operations. The levels radioactive contamination of liquid waste and environment samples were determined in the laboratory of PA «Mayak» from 1949 up to the end of the 1950s as:

-total alpha-activity, Ci
-total beta-activity, Ci
-total gamma-activity, (gram - equivalent Radium).

The methods for determination of total-beta, strontium, cesium, rare-earth element, zirconium/niobium, and ruthenium have been documented. The procedures are described in the report to be made available at the April SRG meeting.

The Techa River has distinct hydrological regimes. There are periods of high water in spring, low water in late summer, and freezing (with no flow at all in some locations) in winter. During the spring high water, contamination may move into and out of marshy areas. The chemical form of the dominant radionuclides has also been evaluated for each of these types of transport period. Measurements describing each of these regimes have been made and recovered. In addition, the characteristics of the river flow changed, first in 1956, and again in 1964, with the addition of Reservoirs 11 and 12 on the upper reaches of the Techa River. All of this information is being compiled for use with the river model. Summaries of the various historical measurements will be available in the report to be made available at the April SRG meeting.

Areas for SRG Discussion

The entire effort of Project 1.3 is exploratory, to determine if available environmental information can be used to reconstruct the Mayak source term to the Techa River. However, there are several areas of ongoing research that could be profitably discussed with the Scientific Review Groups.

Data Quality Objectives This topic is essentially the question of determination of whether the pilot study results deserve expansion into a full-scale project. How good must the initial results be to support the decision to continue efforts beyond the first year? The preliminary results of Phase 2 of this years' efforts are encouraging; they will be used as the basis for the discussion.

Uncertainty Analysis Techniques The source term estimates are based on fitting historical data to a relatively simple mass-balance model. The fitting parameters are determined through numerical optimization procedures. What is the best way to parameterize the uncertainty of the resulting source term estimates?

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